

# Non-linear electronic transport and anomalous resistance fluctuations in the stripes state of $La_2NiO_{4.14}$

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We report on electronic transport measurements in  $La_2NiO_{4.14}$ . Non-Ohmic and hysteretic  $V(I)$  curves are measured for  $T \lesssim 220$  K. Large and non Gaussian resistance fluctuations can be observed, with strong cooling rate dependence. During a slow cooling, the resistance reaches plateaus and then suddenly jumps for  $T \lesssim 100$  K, evidencing a macroscopic freezing of the charges. Anti-correlation between time-series of orthogonal resistances is also observed. These results are discussed in the framework of the stripes state scenario.

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In electronically doped Mott insulators, Coulomb interactions and antiferromagnetic interactions between magnetic ions favor localized charges, giving an insulating tendency. On the contrary, the kinetic energy of doped holes tends to delocalize the charges. The compromise has been shown to result in a charged stripes state, where charge domains are between insulating antiferromagnetic regions [1]. This state is though to be realized in layered nickelates and cuprates. Nevertheless, their transport properties exhibit a major difference. When moderately doped with holes, cuprates can become metal-like ("bad metal") and even superconducting, whereas nickelates exhibit a semiconducting temperature variation up to large hole doping. Extensive neutron scattering experiments have given compelling evidence of the stripes scenario in  $La_{2-x}Sr_xNiO_{4+\delta}$  [2]. Generally, "static" stripes are observed, in agreement with the insulating nature of the nickelates [3]. The stripes state can also be modeled as an electronic liquid phase, using some analogy with the liquid crystals [4]. In particular, the conducting properties of cuprates could be allowed by large transversal fluctuations in a stripe liquid [4]. Recent inelastic neutron scattering experiments seem to

rule out the relevance of static stripes in  $YBa_2Cu_3O_{7-\delta}$  (YBCO) [5], a canonical cuprate, but leave a place for such fluctuating stripes [6].

Since a stripes state is a non uniform 2D electronic system, unconventional transport properties are expected [7]. It is well known that non-linear transport is observed in charges density waves (CDW) systems. CDW can be pinned by the quenched disorder of the sample. Their depinning, above a threshold electric field, leads to a decrease of the differential resistivity [8]. Equivalent features, i.e. the depinning and the associated collective motion, have been predicted for the stripes [9]. Interestingly, it allows some analogies between the stripes dynamics problem and the more general problem of elastic manifolds in quenched random media, generally used for studying the vortex lattices and the CDW dynamics. Unfortunately, the experimental situation is far from being clear in the stripes case. Transport non-linearities has been observed in the charge ordered state of  $La_{2-x}Sr_xNiO_{4+\delta}$  [10], but overheating effects can nuance the interpretation of the data [11, 12]. The attempts to measure non-linearities in the transport properties have been unsuccessful in  $La_{2-x}Sr_xCuO_4$ , despite a detailed study [11]. A possible reason could be that the disorder is not very effective to pin a liquid-like state. As a consequence, the threshold electric field can be too large and not measurable. From another hand, resistance noise due to fluctuations of charges domains can be expected [13], allowing a measure of pertinent properties even at low biasing current. Recently, Bonetti *et al* have observed telegraph-like resistance jumps in the normal state of YBCO nanowires [14], in agreement with the existence of such fluctuating domains. In YBCO, the existence of stripes is still controversial [5]. It is thus important to know if some related effects can be observed in a doped nickelate. This is one of the goal of this experiment.

The sample is a single crystal of  $La_2NiO_{4+\delta}$  with dimensions of  $4 \times 1.9 \times 0.5 \text{ mm}^3$ . The crystal was grown by image furnace. Our sample has an excess of oxygen corresponding to  $\delta = 0.14 \pm 0.01$ , as determined by thermogravimetric analysis. This is very close to the ideal oxygen excess of  $\delta = 2/15$  of the interstitial structure [15, 16]. The quality of the sample has been controlled by EDS, TEM and it was oriented by Laue diffraction technique. The contacts were made with a wire bonding system, connecting Aluminium wires on evaporated gold pads separated by  $150 \mu m$ , in a four probes geometry. In a sample of the same composition, neutron scattering studies have shown that charges order below an ill-defined temperature  $T \gtrsim 220 \text{ K}$ . They have also evidenced that a clear jump in

the intensity of the charge superlattice peak occurs when cooling through  $T_m \approx 110\text{ K}$ , associated with the temperature of spin ordering [15]. Several lock-in transitions of the wave vector of stripe order were also observed for  $T \leq T_m$ . The positions of the observed peaks indicate that the stripes run diagonally through the  $NiO_2$  layers. In our geometry, this is perpendicular to the direction of the transport current (see inset Fig.1).

When dealing with transport non-linearities, a major problem is the overheating due to the Joule effect. In oxides, the limiting points are the low thermal conductivity  $\kappa_{th}$  ( $\kappa_{th}$  is about few  $W/m/K$  for  $T = 100\text{-}300\text{ K}$ ), and the semiconducting temperature variation of the resistance. A maximum transport current  $I^*$  can be applied before reaching a relevant Joule heating. When the applied current is higher than  $I^*$ , a temperature gradient appears between the sample middle and the surfaces, and a notable decrease of the differential resistance is observed [7, 12]. Unfortunately, this effect mimics the depinning of a charge ordered state. To be more quantitative, the  $R(T)$  curve can be fitted by  $R = R_0 \exp(-\beta(T/T_0 - 1))$ , in order to have a simple analytic solution when solving the thermal equations. In this case [12],  $I^* = (2W)(2\kappa_{th}T_0/\beta\rho_0)^{1/2}$ . For  $La_{2-x}Sr_xNiO_4$ ,  $\kappa_{th}$  changes slightly from 300 to 60  $K$  (about 0.03 to 0.2  $W/K/cm$  for the  $x = 0$  compound, [17]). The calculation leads to  $I^* \approx 1\text{ mA}$  for  $T_0 = 60\text{ K}$  ( $\rho_0 = 54640\text{ }\Omega.cm$ ,  $\beta = 6.2$ ), giving here  $E^* \approx 10^4\text{ V/cm}$ . This is close to the values reported in [10]. In our sample, we observe a decrease of differential resistance ( $dV/dI$ ) for values systematically lower than, but reasonably close to, this approximation (not shown). We conclude that the decrease of ( $dV/dI$ ) at high current is likely a heating effect in our sample. Finally, to probe intrinsic processes,  $I \lesssim 500\text{ }\mu A$  was employed for the  $V(I)$  measurements, and all the noise and time series measurements have been performed with a "safe" current ( $I \approx 1\text{ }\mu A$  at high temperature and  $I \approx 50\text{-}500\text{ nA}$  for  $T \lesssim 110\text{ K}$ ).

For  $I \lesssim 1\text{ }\mu A$ , the resistance is found linear in the temperature range  $T = 60\text{-}300\text{ K}$  (the lowest temperatures were not studied due to the too large resistance of the sample). The resistivity increases strongly when the temperature decreases, as previously reported [18], and follows a functional form  $R_0 \exp(T^*/T)^\mu$  (Fig.1). From the Zhabrodskii plot, the plot of  $\ln(-\partial(\ln R)/\partial(\ln T))$  as function of  $\ln T$ , two different exponent  $\mu$  are evidenced (not shown): For  $300\text{ K} \gtrsim T \gtrsim 110\text{ K}$ ,  $\mu = 0.85 \pm 0.05$  (close to a "soft gap"), and for  $110\text{ K} \gtrsim T \gtrsim 60\text{ K}$ ,  $\mu = 0.59 \pm 0.07$  (close to a "variable range hopping" (VRH)). In the "soft gap" regime, a small peak of the activation energy can be observed at  $T \approx 210\text{ K}$ . These exponents are *not* in very good agreement with what is expected in the case of

conventional VRH or activated modeling. This is likely due to the intrinsic inhomogeneity of the electronic ground state, which should invalidate conventional treatment. One can only note that the temperatures of cross-over are close to the ordering temperatures extracted from neutron scattering experiments [15].

In Fig.2, the  $V(I)$  curves are shown. They are measured at different temperatures in the "safe" current regime previously discussed. Both non-ohmicity and hysteresis are observed for  $T \lesssim 220$  K. This can be caused by a stripes depinning in the presence of quenched disorder [19]. The threshold currents are in the range 1-100  $\mu A$ . This corresponds to rather low electric field of  $10^{-1}$ -10 V/cm, close to what is expected for a conventional CDW depinning [8]. This electric field exhibits an activated temperature dependence  $E_0 \exp(T^*/T)$ , with energy barrier  $T^* \approx 450$  K (not shown). This is characteristic of a thermal assisted process, and agrees also with some CDW depinning models [8]. Anyway, stripes or CDW depinning favors conductivity [9], but the system becomes here *more* resistive at high current. After several cycling, we observed that the  $V(I)$  curve tends to be linear, meaning that the high resistance state is the most stable one. We conclude that we do not observe a genuine stripes depinning. A possibility is that the resistance switching marks a transition to a stable state where charges are more localized. The  $V(I)$  measurements have been made after cooling the sample from 300 K with a rate of about 0.5 K/min. A systematic cooling rate dependence of the  $V(I)$  curves has not been performed.

We have shown that the  $R(T)$ , measured under a moderate cooling rate condition (2 K/min), is smooth and without obvious characteristics (see Fig.1). More insights into the existence of metastable states come from the study of resistance fluctuations. From an experimental point of view, we have measured resistance time-series  $R(t)$ , at a constant temperature after cooling the sample, during long times (1-2 hours). From these time-series, the mean square value  $\overline{(\delta R^*)^2}$  can be calculated [20]. The temperature variation of  $\delta R^*$  is shown in Fig.3. Typically, three regimes can be defined and will be discussed below.

The first regime is for  $240$  K  $\lesssim T \lesssim 300$  K: the resistance is observed constant as function of the time whatever the cooling rate. The second regime is for  $200$  K  $\lesssim T \lesssim 240$  K. After a fast cooling of the sample from  $T = 300$  K, a slight and continuous decrease of the resistance as a function of the time can be observed (not shown). After turning off the current during 1000sec and then restoring it, the relaxation restarts as if the current was not turned off. This shows that the system has intrinsically evolved during the time. This

effect can be the counterpart of the very low kinetic of the growth of the interstitial oxygen ordering after quenching the sample, such as reported from neutron diffraction study [21]. If the sample is cooled from  $T = 300\text{ K}$  to the same temperature, but with a low or moderate cooling rate ( $\lesssim 1\text{ K/min}$ ), the time series is smooth and stationary and does not contribute to the excess noise.

For  $T \lesssim 200\text{ K}$ , a third and more interesting regime is observed.  $\delta R^*(T)$  exhibits large peaks for several temperature values (Fig. 3). These peaks correspond to non-stationary time-series with large resistance steps, as evidenced in the inset of the Fig.3. The typical lifetime of one resistance state is several hundred of seconds. After a very long time (up to 2 hours), the resistance tends to a quasi-stationary behaviour, where only two-states switching can be rarely evidenced. This is quite different than the telegraphic-like noise observed in YBCO, this latter being associated to random fluctuations of charges/stripes domains [14]. In contrast, the non-stationarity observed here implies a more complicated kinetic in  $La_2NiO_{4.14}$ . The time-series look like crackling noise [22], i.e. exhibit discrete events with different sizes, as shown in Fig.4. The histogram of the size of resistance steps  $\Delta R$  has power-law dependence  $\Delta R \propto R^{-s}$  with  $s \approx 1.3$ , in general agreement with avalanche exponents found in other systems [23] (note that, in our case, the restricted number of events does not allow to prove rigorously a power-law). For the well documented case of magnetic avalanches, the domain wall motion in a disordered landscape should be responsible for the avalanche behavior [24]. In the framework of stripes dynamics, this implies that disorder is relevant in our doped nickelate, in agreement with [25]. We have also observed that the typical time scale of the resistance fluctuations is dependent of the cooling rate, suggesting some glassy dynamics. This effect is more pronounced at low temperature. In Fig. 5, the resistance time-traces are shown at  $T = 60\text{ K}$ , after a fast cooling ( $5\text{ K/min}$ ) or a slow cooling ( $\lesssim 0.5\text{ K/min}$ ) from  $T = 300\text{ K}$ . We have noted that the general features are well reproduced after identical thermal histories. The slow cooling favors more probable resistance steps, proving that they do not come from a frozen disorder due to temperature quenching. For such a slow cooling, a very peculiar behavior can be observed. As shown in Fig. 6, when measured continuously during the cooling, the resistance tends to saturate and then suddenly jumps for some temperatures  $T \lesssim 100\text{ K}$ . In the same temperature range, commensuration effects have been observed between the stripes spacing and the lattice spacing [15]. This shows strong coupling with the lattice potential, and besides, sensitivity

to disorder [25]. The plateaus in the  $R(T)$  evidence a freezing of the charges at a macroscopic scale due to this strong coupling. We think that a slow cooling allows a strongest efficiency of the disorder landscape sampled by the system.

In addition, we have observed anticorrelation between resistance fluctuations measured along and perpendicular to the current flow (see Fig.7). This anticorrelation is particularly clear for the large resistance steps. In [13], it has been discussed that a nematic long range order, such as present in a stripes phase, is preserved only locally when it is coupled with the quenched disorder. The result is a 2D distribution of stripes patches whose motion or reorganization can give rise to crackling noise and to anticorrelation in resistance noise [13]. At higher temperatures ( $T \gtrsim 100\text{ K}$ ), this anticorrelation becomes unmeasurable. One can note that  $T \approx 110\text{ K}$  is a characteristic temperature in neutron scattering experiment, marked by a sudden and strong increase of the neutron intensity, and associated with the stripes ordering [15]. Our measurement of anticorrelation seems consistent with a global picture of an ill-defined stripe order at high temperature, which freezes, coupled to the quenched disorder, for  $T \lesssim 100\text{ K}$ .

To conclude, non-linear electronic transport and anomalous resistance time-series have been measured in  $La_2NiO_{4.14}$ . Unlike in YBCO where relatively quiet and two states fluctuations have been measured [14], strong non-stationary effects are observed in this nickelate. We do not observe the genuine depinning of a charge ordered state, but rather a reorganization of metastable states after cooling the sample. If the link we make between these features and the stripes state is correct, this latter shows a very slow and constrained kinetic, likely due to a strong coupling with the sample disorder.

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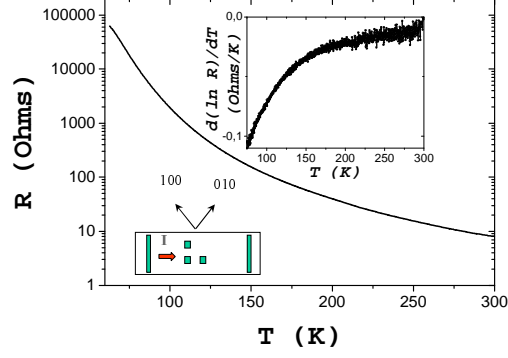


FIG. 1: (Color online) Low current linear resistance of the  $La_2NiO_{4.14}$  crystal as function of the temperature (cooling rate  $2\text{ K/min}$ ,  $I = 1\text{ }\mu\text{A}$ ). In the higher inset is shown the derivative of the resistance logarithm as a function of the temperature. In the lower inset is shown the schematic view of the sample with the contact pads. The cristallographic directions are also reported.  $R$  stands for the resistance along the transport current  $I$ ,  $R_{perp}$  is perpendicular to  $I$ .

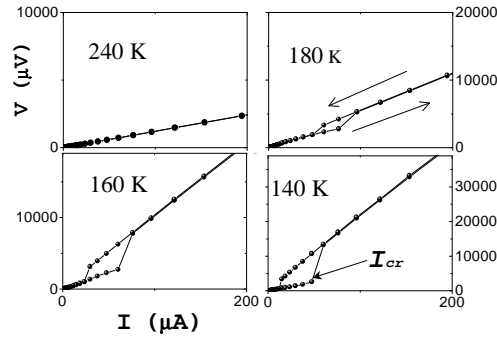


FIG. 2:  $V(I)$  curves for  $T = 240, 180, 160$  and  $140\text{ K}$ .  $I_{cr}$  corresponds the current where a voltage jump can be observed when the current is increased. The cooling rate before each acquisition is  $0.5\text{ K/min}$ .

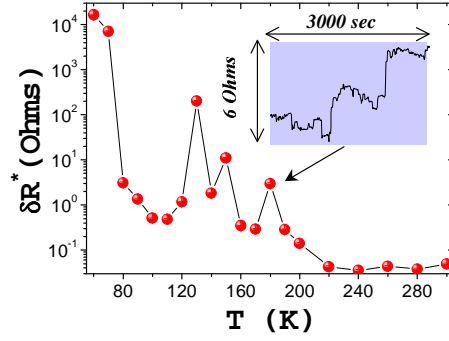


FIG. 3: (Color online) Resistance noise  $\delta R^*$  (integrated over a  $10^{-2}$ - $10$  Hz bandwidth) as function of the temperature ( $I = 1 \mu A$ ). In the inset, a typical resistance time-series is shown ( $T = 180$  K). Between the noise peaks, the time-series are stationary and without excess noise. The typical cooling rate is  $0.5 K/min$

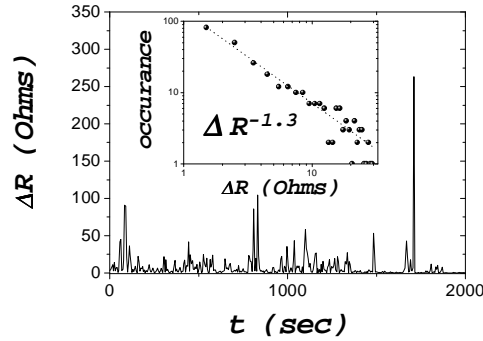


FIG. 4: Size of the resistance steps  $\Delta R$  as function of the acquisition time ( $T = 130$  K,  $I = 0.5 \mu A$ ). In the inset is shown the histogram of  $\Delta R$  and the fit using a functional form  $\Delta R^{-1.3}$ .

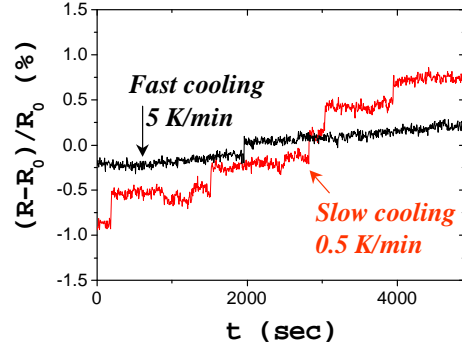


FIG. 5: (Color online) Time-traces of the resistance at  $T = 60$  K, after a slow cooling or a fast cooling ( $I = 0.1 \mu A$ ). Note the increase of the rate of resistance steps after the slow cooling.

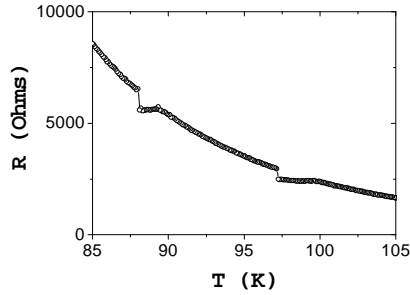


FIG. 6: Resistance as a function of the temperature during a slow continuous cooling (rate=  $0.5$  K/min,  $I = 0.5 \mu A$ ). Note the two plateaus, which are followed by jumps to the "normal" resistance value.

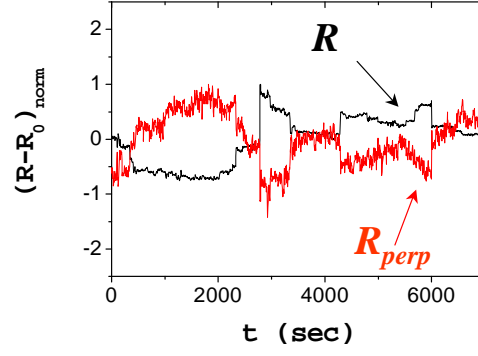


FIG. 7: (Color online) Time series of resistance in orthogonal directions (rate=  $0.1 \text{ K/min}$ ,  $T = 60 \text{ K}$ ,  $I = 1 \text{ } \mu\text{A}$ ).  $R_{\text{perp}}$  is the resistance perpendicular to the direction of the current. Each trace has been normalized. Strong anticorrelation can be observed between the two time-series.